Crack propagation through phase separated glasses: effect of the characteristic size of disorder

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We perform fracture experiments on nanoscale phase separated glasses and measure crack surface roughness by atomic force microscopy. The ability of tuning the phase domain size by thermal treatment allows us to test thoroughly the predictions of crack font depinning models about the scaling properties of crack surface roughness. It appears that in the range of validity of these depinning models developed for the fracture of brittle materials, our experimental results show a quantitative agreement with theoretical predictions: beyond the characteristic size of disorder, the roughness of crack surfaces obeys the logarithmic scaling early predicted by Ramanathan, Ertas and Fisher[1].

Ever since an early paper by Mandelbrot *et al* [2] about the fractal character of crack surfaces, a growing interest has developed in the understanding of fracture of heterogeneous materials[3, 4, 5]. In particular many studies[2, 4, 6, 7, 8, 9, 10, 11, 12] have dealt with the statistical characterization of both crack front geometry and crack surface roughness. It appears that, over a broad spectrum of length scales, these objects obey a self-affine symmetry: they are left statistically invariant rescaling with different ratios depending on the direction. More specifically, self-affinity implies that the typical height differences Δh along the surface scales as $\Delta h \propto \Delta x^{\zeta}$ when measured over a distance Δx, ζ being a real parameter called the roughness, or Hurst, exponent. Early studies proposed that the roughness of various fractured materials, independently of their ductile[6] or brittle[7] nature, could be described over a wide range of scales with a unique value of the roughness exponent $\zeta \simeq 0.8$.

This apparent universality motivated the development of models[1, 13] based on the depinning of an elastic line through a random landscape[14] and the description of long range elastic interactions along a rough crack front[15, 16]. A crack surface can indeed be considered as the trace left by a crack front propagating through a disordered material[17] (see Fig. 1a for a representation of crack front propagation). In such a description, a particular site pinned by a microscopic tough zone may overcome the obstacle with the help of the additional elastic force due to the straining of the structure around the obstacle. This competition between a roughening effect due to the quenched random environment and a smoothing effect due to elastic interactions along the front leads to a rich phenomenology. When the Stress Intensity Factor K remains below a certain threshold, K^* the front can only propagate over a finite length and then stops; above threshold, the front can move at a finite, but highly fluctuating, velocity. Moreover, this transition appears to be critical, with a collection of critical exponents (among which the roughness exponent of the elastic front) ruling its behavior close to threshold, with the strong property of universality coming naturally with this observation.

These depinning models thus intend to characterize the associated critical transition and to capture the geometry and the intermittent dynamics of the crack front. The case of interfacial plane cracks was first developed[13] to study the in-plane roughness (*i.e.* in the direction of propagation) of the front in the propagation plane. The more complex case of a 3D crack front was then discussed[1], allowing the study of the out-of-plane roughness (*i.e.* in the direction normal to the mean plane of propagation) responsible for the fracture surface roughness.

Though qualitatively correct, the results appeared to be quantitatively rather disappointing since these models predict only a logarithmic scaling of the crack surface's out-of-plane roughness [1], *i.e.* the roughness exponent predicted by the depinning model was $\zeta^{dep} = 0$ (here and in the following the notation ζ^{dep} will refer to the exponents predicted by the depinning models while the notation ζ will refer to experimental estimates) whereas the experimental estimate was $\zeta \simeq 0.8$. Even in the simpler case of plane crack propagation, the roughness exponent characterising the in-plane roughness of the crack front was found to be significantly lower in models ($\zeta_H^{dep} \simeq 0.4$) [18, 19] than in experiments ($\zeta_H \simeq 0.55$) [8].

We argue here within that the main reason for the apparent discrepancy between experimental results and scaling predictions of depinning models stems from the dubious status in most experiments of a key hypothesis of the models: a clear separation between the scale of disorder and the scale of measurement. To test this argument, we present results of fracture experiments performed on a series of model heterogeneous brittle materials: phase separated glasses[20]. Thermal treatments of various durations allowed the fine tuning of phase domain sizes. Phase separated glasses thus differ only by one single parameter, the lengthscale of their microstructure, with all other material properties

Heat treatment duration	4h	16h	64h
Phase domain size d	28.2 ± 2.5 nm	56.3 ± 9.3 nm	92.2 ± 9.4nm

TABLE I: Size d of phase domains (estimated by the correlation length at half height) *vs* duration of thermal treatment at 650°C. See Ref. [20] for quantitative details about the kinetics of phase separation on these glasses.

remaining identical. Such an experimental procedure thus allows us to study the dependence of the spatial extent of the scaling regime on the characteristic size of the internal disorder.

Phase separated glasses have been prepared from an alkali borosilicate glass of composition (in weight): SiO_2 70%, B_2O_3 25%, Na_2O 2.5% and K_2O 2.5%. The raw materials were mixed together and melted at 1550°C and the melt was then refined over 2 hours in order to obtain a homogeneous glass; after quenching, the glass was annealed for 1h at 630°C in order to relax the internal thermal stresses. At this stage, phase separation had already initiated, attested to by the slightly opalescent character of the glass. The major phase is near-pure silica while the minor phase concentrates other constituents. Heat treatments at 650°C over increasing durations (4h, 16h, 64h) were then applied to increase more and more the size of phase domains as shown in Fig. 2 and summarised in Table I. It was thus possible to prepare glasses with controlled phase domain sizes, ranging from around 20 to 100 nm. Further details concerning the preparation and the characterisation of the glass samples can be found in Ref. [20].

Stable dynamic fracture experiments on these heterogeneous materials were performed by using a DCDC set-up (Double Cleavage Drilled Compression) (see Fig. 1-Left) and a statistical analysis of the fracture surface roughness carried out from Atomic Force Microscopy (AFM) images (see Fig. 1-Center for the principle and Fig. 1-Right for an AFM image of roughness masurements obtained on a fracture surface). From the above mentioned glass, parallepipedic shaped samples (5mm×5mm×25mm) with a central hole 1mm in diameter have then been prepared. Stable cracks were then propagated in these by using DCDC mechanical test. Roughness measurements, coming from height images (TM-AFM) were then immediately performed in areas lying between two crack arrest lines. The measurements are thus performed in the immediate vicinity of the propagation threshold: $K \approx K^*$.

The surface morphology of all samples has been characterised by AFM height measurement in tapping mode (TM-AFM), using a Nanoscope III A from Digital Instruments with Al coated tip (BudgetSensors - model BS-Tap 300 Al). The Al coating thickness used was 30nm, the resonant frequency 300 kHz and the stiffness constant 40N/m. Images were recorded at a scan frequency between 0.8 and 1Hz for a resolution of 512×512 pixels. For each sample (i.e. for each thermal treatment conditions), at least 3 AFM images were performed for 4 different scan areas ranging between 500×500 nm^2 and 8×8 μm^2.

Beyond its imaging ability, AFM has recently been used as a truly quantitative tool to study glass surfaces. It is, for example, possible to obtain a quantitative validation of the description of fused glass surfaces by frozen capillary waves[21] or of the kinetics of phase separation in glasses[20]. In both cases, not only are the expected scaling regimes recovered but the prefactors of the scaling laws can also be extracted and shown to be consistent with the associated physical parameters (diffusivity, interface tension). In the present case of fracture surfaces, it appears that one can identify an apparent scaling regime up to the size of the heterogeneities (Fig. 3a). This result is consistent with the standard analysis even if the scaling range is limited. More interestingly, beyond the size of heterogeneities, one observes (Fig. 3b) a clear logarithmic scaling as predicted by the depinning model early proposed by Ramanathan *et al* [1, 22].

More specifically Fig. 3 represents the evolution of height differences $\Delta h(\Delta x) = \left(\langle [h(x+\Delta x) - h(x)]^2 \rangle_x\right)^{1/2}$ measured on a scale Δx on the crack surface of each of the three types of sample. The results could be collapsed onto a master curve after rescaling by the phase domain size d along the x-axis and by the typical height difference $\Delta h(d)$ along the $y-$axis. When data are displayed on a log-log scale (Fig. 3a), an apparent power law behaviour of exponent $\zeta = 0.8$ is identified, extending over one decade up to the size of heterogeneity. Such a scaling law *below* the size of heterogeneities, if genuine, could not be explained in the framework of depinning models which predict scaling regimes only *above* the typical scale of disorder. However if data are now represented on a simple linear-log scale, we obtain above the characteristic scale of the heterogeneities excellent agreement between our results and the logarithmic scaling predicted by depinning models. Such a data collapse after rescaling by the characteristic size of disorder ξ_0 is fully consistent with a description of fracture propagation as a critical transition. In such models the scaling regime is indeed expected to hold in the spatial range $[\xi_0, \xi]$ where ζ_0 is the characteristic size of the toughness disorder $\xi \propto \xi_0 |K^* - K|^{-\nu}$ is the correlation length, diverging as a power law of exponent ν when the stress intensity factor K approaches the critical threshold K^* (which is nothing here but the effective toughness of the material) and is directly proportional to ξ_0. Here, propagation is performed close to threshold and the scaling regime is thus expected to be wide enough to be easily observed. Note that in case of a strongly overdriven crack, $K \gg K^*$, dynamic

instabilities set in and a roughness of entirely different nature is expected to appear. Depinning models thus seem to give an excellent account of the scaling of the roughness of crack surfaces obtained from the quasistatic fracture of brittle heterogeneous materials such as the phase separated glasses used in the present study.

The present results thus may contribute to clarify the current debate about fracture and universality. In view of the persistent discrepancy between experimental measurements and model predictions, efforts have indeed been made to either improve these depinning models[23, 24] or look for alternative scenarios for fracture propagation[11, 25]. Higher order terms were taken into account in the depinning model[23] but apparently did not change the scaling behaviour[26]. High velocity propagation was discussed[24] as a possible mechanism to anneal the frozen toughness noise in the equation describing the front propagation. The interplay between local damage and crack propagation was proposed but led to controversy at both the theoretical[27, 28, 29, 30] and the experimental[31, 32] levels.

Moreover, in light of the variety of experimental results, the initial claim of universality itself may need to be tempered. Beyond the variability of sacling exponents estimated experimentally, when measurements are performed on materials with a well defined micro-structure such as glass[11], sintered glass beads[12] or some cast aluminium alloys[10], it appears that either the scaling range is very limited or that it does not extend beyond the grain size whereas a truly universal scaling regime should extend beyond the characteristic size of heterogeneities.

Within this context, the results of the present study suggest a simple scenario. When considering the stable propagation of *brittle* heterogeneous samples, depinning models can account quantitatively for the scaling of crack surface roughness *beyond* the scale of heterogenities. This means in particular that we need to clearly separate the spatial scales within which the material can be considered as brittle (beyond the size of the "process zone" where dissipative mechanism associated with crack propagation take place) from the range of scales within which damage or ductility take place. Depinning models will not give any predictions below the size of the process zone. It will thus be necessary to resort to alternative models (stress-weighted percolation[28], random fuse models[5]) to account for fracture propagation in *ductile* heterogeneous materials.

Note again that in the present framework of fracture, the notion of disorder is rather subtle. The fluctuating quantity to consider is actually the toughness of the material. However the toughness disorder is not a direct reflection of the structural disorder but need be "convoluted" at the scale of the process zone. Two lengthscales have thus to be considered, the size of the process zone and the characteristic size of the structural disorder. In the present case, glasses being very brittle, the typical size of the proces zone does not overcome a few nanometers, below the size of the phase domains and the characateristic scale of the toughness disorder can be safely identified with the size of the structural disorder.

The common experimental observation of a wide scaling regime on fracture surfaces with roughness exponents differing from the depinning predictions may therefore result from the non-respect of the hypothesis of depinning models: scale separation between toughness disorder and experiments or perfect brittleness. This may be the case for ductile materials for which the size of the process zone lies in the spatial range of roughness measurements or very heterogeneous materials presenting heterogeneities over a wide range of scales. In particular the "classical" scaling regime characterized by a roughness exponent $\zeta \simeq 0.8$ may correspond to the existence of additional microscopic mechanisms (*e.g.* damage or other non-linear behaviour) over a more limited range of scales. The present results also suggest that experiments displaying a scaling regime only up to the characteristic size of disorder may be revisited to test whether they exhibit the logaritmic scaling of roughness expected beyond that characteristic size.

This first quantitative validation of the depinning scenario to describe brittle crack propagation through heterogeneous materials thus provides a framework to extend the use of critical transitions concepts to fracture, for instance in order to better predict fluctuations and homegeneous behaviour from nano- to macro-scale[33].

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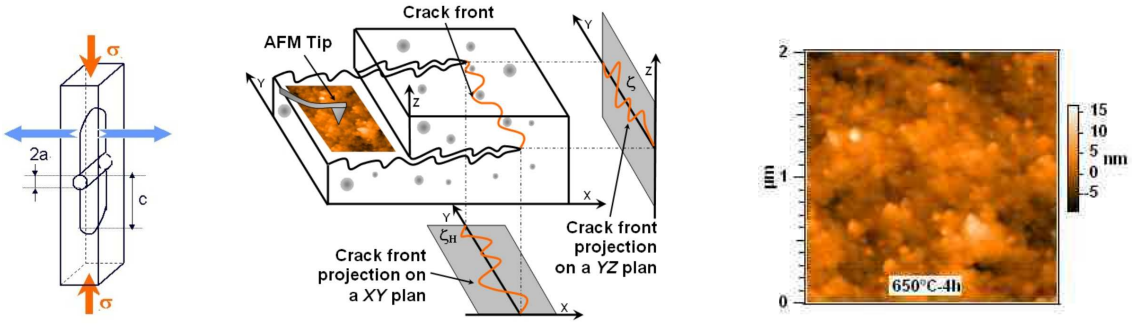

FIG. 1: **Experimental methods.** Left: geometry of the DCDC mechanical test. Two longitudinal cracks propagate from the cylindrical hole. Center: sketch of a propagating crack front pinned by heterogeneities (here figured as grey dots). The front develops a roughness both in the out-of-plane direction, visible on the $y-z$ plane projection and in the direction of propagation, visible on the $x-y$ plane projection. When scaling invariant, this roughness is characterized by an exponent ζ in the out-of-plane direction and an exponent ζ_H in the direction of propagation. AFM measurements are performed on the crack surface. Right: AFM image of a surface obtained after fracture of a glass sample annealed at 650°C over 4h.

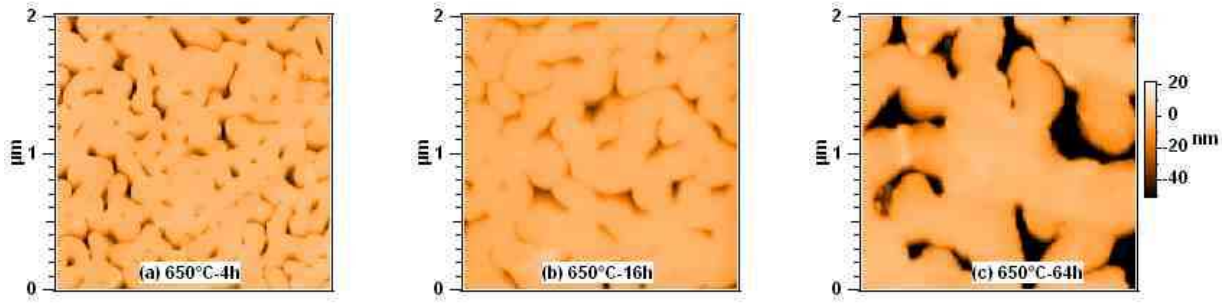

FIG. 2: **AFM images obtained after acid etching on glass samples annealed at 650°C over 4h, 16h and 64h respectively**. Under annealing, alkali borosilicate separate into a near pure silica phase and another phase concentrating all other components. The latter phase can be eliminated by acid etching, the height contrast obtained on AFM images thus reveals the phase contrast (see also Ref. [20]).

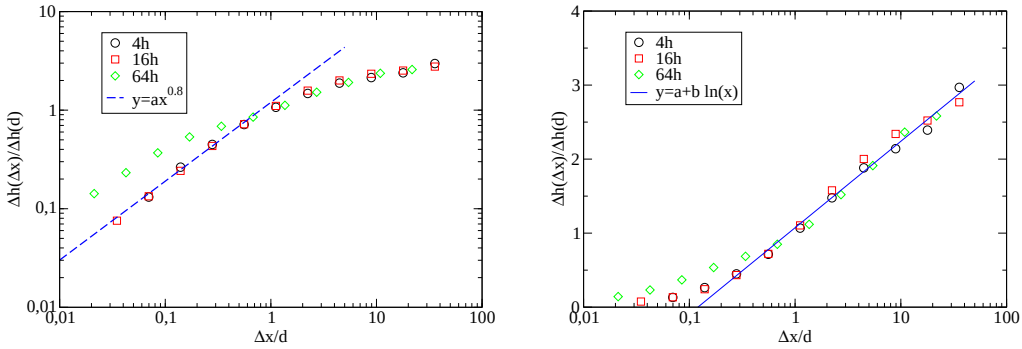

FIG. 3: **Roughness of fracture surfaces after rescaling by the phase domain size d along the $x-$axis and the typical roughness $\Delta h(d)$ at size d along the $y-$axis**. Left: in log-log scale an apparent self-affine regime with exponent $\zeta = 0.8$ may be identified up to the the size of the phase domains. An indicative power law of exponent 0.8 is displayed. Right: in linear-log scale, a simple logarithmic regime appears to describe the surface roughness as predicted by depinning models. The straight line corresponds to a logarithmic fit performed on the 3 sets of data for lengths larger than the domain size.